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Global Nuclear Energy Partnership Waste Treatment Baseline

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Abstract – *The Global Nuclear Energy Partnership program (GNEP) is designed to demonstrate a proliferation-resistant and sustainable integrated nuclear fuel cycle that can be commercialized and used internationally. Alternative stabilization concepts for byproducts and waste streams generated by fuel recycling processes were evaluated and a baseline set of waste forms was recommended for the safe disposition of waste streams. Specific waste forms are recommended based on the demonstrated or expected commercial practicability and technical maturity of the processes needed to make the waste forms, and expected performance of the waste form materials when disposed. Significant issues remain in developing technologies to process some of the wastes into the recommended waste forms, and a detailed analysis of technology readiness may lead to the choice of a different waste form than what is recommended herein. Evolving regulations could also affect the selection of waste forms.*

INTRODUCTION

The United States Department of Energy (DOE) Global Nuclear Energy Partnership (GNEP) program is designed to demonstrate a proliferation-resistant integrated nuclear fuel cycle.[1] This fuel cycle consumes transuranic (TRU) elements and supports growth of carbon-free international nuclear energy markets. Building on the knowledge gained over the last 60 years of nuclear science and engineering, the proposed recycling system is not only more sustainable than prior concepts, it will generate less long-lived waste and reduce the thermal load and impacts of long-lived radionclides on a geologic repository. Key to successful implementation of a closed nuclear fuel cycle in the United States is establishing an Integrated Waste Management Strategy (IWMS) to provide for routine disposition of all byproduct and waste streams. Documents published in February and May 2007 summarized the wastes expected from aqueous and electrochemical fuel recycling[2] and the technical and policy issues affecting their safe disposition.[3] This paper provides the technical bases supporting the selection of recommended waste forms for all the waste streams, based on anticipated commercial practicability, technical maturity, and waste form performance. Each waste form discussion also identifies the remaining uncertainties and data needs regarding the manufacture and performance of these waste forms that should be addressed in on-going research activities.

WASTE STREAMS

The Advanced Fuel Cycle Initiative (AFCI), which was the predecessor program to GNEP, sponsored extensive R&D efforts related to aqueous-based and electrochemical separations processes for recycling thermal reactor fuel, including light water reactor (LWR) and fast reactor (FR) spent nuclear fuel (SNF). The fuel cycle proposed in GNEP consumes TRU elements and supports growth of carbon-free international nuclear energy markets. Building on the nuclear science and engineering knowledge gained over the last 60 years, the proposed recycling system is not only more sustainable than prior concepts, it is also intended to generate less long-lived waste per equivalent energy production than the current once through fuel cycle. Waste management is significantly more protective of the environment because the remaining radionuclides will be in more protective waste forms than directly disposed SNF.

The research conducted through AFCI was primarily focused on developing understanding of the chemistry and performance of aqueous and electrochemical separations to recover uranium from spent fuel. This has resulted in a suite of aqueous processes based on solvent-liquid extraction known as UREX+1a, and electrochemical partitioning using electrical potential applied across a molten-salt bath , referred to as Echem. This research has also resulted in an understanding of the basic characteristics of the waste streams that are expected from the separations activities. The

primary product, byproduct, and waste streams considered in the IWMS and their planned disposition routes are excerpted from the IWMS Issues Report[3] and updated here in Tables 1 and 2. Several of the waste forms described in the tables are experimental and designed to complement the advanced separations in UREX+1a to manage heat and to minimize waste volume and long-term radiotoxicity sent to a geologic repository.

TABLE 1. Waste Streams and GNEP Baseline Disposition for Aqueous (UREX+1a) Reprocessing.

Aqueous Process Waste Stream	Stream Description/Derivation	Current Baseline Disposition
Assembly hardware primarily stainless steel (SS)	Spacers, endcaps, etc., removed prior to chopping of fuel made of various stainless steels, Inconels, and Zircaloys	Compact for Direct repository disposal, evaluate performance assessment for shallow land burial (SLB) of greater than Class C (GTCC).
Gaseous Products Kr/Xe and ^{3}H	Voloxidation releases Kr/Xe and ^{3}H which are caught on absorber beds	Decay storage of Kr/Xe and ^{3}H followed by SLB of packaged forms as low-level waste (LLW) or release
Iodine, Carbon-14	Sorption of I on silver zeolite, and incorporation of ^{14}C as carbonate	SLB if Class A/B/C LLW, geologic repository of GTCC
Hulls/Cladding (Zr)	Residual metals following fuel dissolution	Disposal as GTCC due to activation or Contamination
Undissolved Solids (UDS)	Sludge from dissolver bottom and clarifier solids containing noble metals and TRU	Combine with other metallic waste for repository disposal
Separated LEU	Oxidation of uranyl nitrate solution from UREX to UO3 or U3O8	Store as national resource material, recycle as fuel, or SLB as LLW
Tc on ion exchange (IX) resin	Acid side ion-exchange of UREX raffinate to be stripped or pyrolyzed and reduced to Tc metal	Combine with other waste for repository disposal
Cs/Sr stream	CCD/PEG[a] or FPEX[b] solvent extraction of UREX raffinate, yields Cs, Sr, barium (Ba), and rubidium (Rb)	Stabilize for long-term (100 to 300 yr) decay storage and disposal in repository, SLB after decay if LLW
TRU stream	Oxidize TRU either Pu/Np and Am/Cm separately or together	Product for FR fuel or target fabrication
Lanthanides and transition metal fission products (FP) streams	TRUEX[c] raffinate and TALSPEAK[d] product combined or separate	Stabilize for repository disposal
Liquid waste (aqueous and organics)	Liquids from several locations in the process, including off-gas treatment, spent solvents, solvent wash, laboratory returns, and other miscellaneous liquids	Stabilize to solids, SLB of oxide as LLW
Miscellaneous Solid low level debris	Spent equipment, PPE, laboratory, and operation solid waste (pipettes, wipes, etc.), after decontamination	Direct SLB as LLW or disposal as GTCC as required

a. CCD/PEG - chlorinated cobalt dicarbollide-polyethylene glycol
b. FPEX - fission product extraction
c. TRUEX - transuranic extraction
d. TALSPEAK - Trivalent Actinide Lanthanide Separations by Phosphorus-reagent Extraction from Aqueous Complexes

TABLE 2. Waste Streams and Baseline Disposition for Echem Processing.		
Echem Process Stream	Stream Description/Derivation	Current Baseline Disposition
Volatile Products (I, Kr/Xe, ^{3}H)	Released during chopping process and electrorefining and caught on absorbers	Decay storage of Kr/Xe and ^{3}H followed by SLB of packaged forms as low-level waste (LLW) or release
Zr/SS cladding and hardware from processing oxide fuels	Hardware is sectioned and removed, Zircaloy cladding is removed from powdered fuel after voloxidation treatment	Direct repository disposal, evaluate performance assessment for SLB of GTCC.
Residual metals and undissolved solids (UDS) from processing metal fuels	Undissolved SS metal cladding stream from electrolytic dissolution includes Tc, Zr, and noble metals	Melt as metal waste form for repository disposal
Residual transition metals and UDS from processing oxide fuels	Undissolved metal waste stream from electrolytic dissolution includes Tc, Zr, and noble metals	Melt as metal waste form for repository disposal
Separated low-enriched uranium (LEU) from either oxide or metal fuel	Deposited on iron cathode as U metal, stripped and heated to remove adherent salts	Store as national resource material, recycle as fuel, or SLB as LLW
U/TRU stream	TRU electrolytically partitioned with some LEU	Product for FR fuel fabrication
Cs/Sr from processing oxide fuel	Capture from salt bath on zeolite, contains Cs, Sr, Ba, and Rb	Stabilize for long-term (100 to 300 yr) decay storage and disposal in repository, SLB after decay if LLW
Lanthanides	Lanthanides electrolytically partitioned	Stabilize for repository disposal
Waste salts with Cs/Sr, FP, iodine and carbon-14 from metal fuel or without Cs/Sr if oxide fuel	Salt bleed stream containing non-reducible salts	Convert to glass-bonded sodalite for repository disposal
Miscellaneous low level solid debris	Spent equipment, PPE, laboratory, and operation solid waste (pipettes, wipes, etc.), after decontamination	Direct SLB as LLW or disposal as GTCC as required

As can be seen in Tables 1 and 2, the GNEP concept is based on advanced separations that make it possible to partition spent fuel into several fractions rather than composite high level waste (HLW), thereby partitioning the radionuclides into groups of common chemistry and to a great extent, common risk. Advanced separations allow greater flexibility in managing the individual waste streams based on duration, type, and magnitude of risk, and to develop specialized waste forms to sequester radionuclides effectively per the IWMS.

Recovery of actinides is a central goal of GNEP to reduce the potential for proliferation, benefit from the fuel value of plutonium, and reduce the long-term radiotoxicity and heat in a geologic repository. Next, partitioning of readily oxidized alkali and alkaline earth elements that are relatively short-lived but generate substantial heat (Cs/Sr) allows creation of a waste form that can be managed to dissipate heat prior to disposal, thereby mitigating heat limit affects on repository capacity. Similarly, very short-lived gaseous radionuclides including tritium and krypton can be captured and allowed to decay in storage prior to eventual disposition. Segregation of the lanthanides allows production of a high-waste loading lanthanide-based glass, thereby reducing volume sent to a repository.

Currently, the baseline waste form for HLW worldwide is borosilicate glass containing waste elements as oxides. Unfortunately several fission product elements have low solubility in glass, which results in low waste loading, and

production of more glass. With the other elements removed, the remaining metallic fission products that have historically limited HLW loading in glass can potentially be alloyed with cladding and undissolved solids in a separate waste form to minimize the total volume.

Matching the waste form to the individual waste stream chemistry allows the disposal system to achieve more optimum waste loading with less heat and radiotoxicity achieving comparable or improved performance. Not only can the waste form be matched to the waste, but the disposal environment can be matched. Some elements are more stable in a low-oxygen reducing environment, while others are more stable as oxides. Thus a more efficient waste management system that uses the most effective waste form and disposal design for each waste is made possible by this proposed change in technology. The following sections discuss the options evaluated for the primary radionuclides separated in GNEP advanced separations.

Reducible Element Waste Streams

For the aqueous process, the reducible element waste streams are the UDS from the fuel dissolution step, dissolved Tc recovered from the UREX product, and transition metal FP recovered from TRUEX. Depending on the conditions that are used, it is expected that at least 75% of the Tc in the fuel will be dissolved in the initial dissolution, and it is assumed that all of the dissolved Tc will be recovered from the UREX solution. The balance of the Tc will be in the UDS. The dissolved fractions of the other transition-metal fission products (FP) that dissolve including Zr, Mo, Ru, Rh, and Pd will be recovered in the TRUEX raffinate. For waste management efficiency, this FP waste stream could potentially be blended into the Tc/UDS-bearing waste form because these transition metals are generally the same elements that make up the UDS.

For the Echem process, the reducible metals including Tc and the transition metals described above are captured together as the metallic wastes that are retained in the anode basket of the electrorefiner.

In addition to metals from the fuel itself, some waste streams will include metals used in processing. One method being studied for recovering pertechnetate from the UREX solution is to evaporate the solution and reduce pertechnetate to the metal by steam reforming. The metallic Tc would then be alloyed with Zr. Another method being studied is the reductive deposition onto iron or other metal substrate (including electrodeposition). This would eliminate the need to evaporate the solution and steam reforming, and would provide the Fe needed to produce the alloyed waste form. If the transition metal fission products are combined with this stream, ferrous sulfamate added to reduce Np(V) to Np(IV) in the solution prior to TRUEX separation are included. This makes iron the dominant metal in the FP-bearing waste solution and in the blended Tc, UDS, and FP streams. The use of reductants other than ferrous sulfamate is being studied to eliminate Fe and S from the waste stream.

In the Echem process, the anode basket used in the electrorefiner is expected to be included with the waste, and would dominate the waste stream composition. The basket is constructed primarily of Type 316 SS and will contribute a significant amount of Fe to the waste stream as well as small amounts of Cr and Ni. It is anticipated that the cladding hulls from the oxide fuels that are treated with either the UREX or Echem processes will be cleaned and disposed of separately from the Tc-bearing metallic wastes. The hulls may be compacted for direct disposal or alloyed. The cladding hulls from metallic fuels will be treated with the Echem process and retained with the Tc-bearing metallic wastes in the anode basket and alloyed with them in a metallic waste form.

Immobilization of the Tc-bearing aqueous waste streams in a multi-phase metal-alloy waste form or a borosilicate glass was evaluated. In the Echem wastes, Tc is combined with massive amounts of other metals and vitrification is not practical. Vitrification was considered because Tc is being immobilized in glass internationally, albeit at very low concentrations. However, vitrification of the GNEP reducible metal wastes is probably not practical because of:

1. restrictive limitations on the amounts of metals that can be tolerated in a melter,
2. volatilization of nonmetallic Tc at vitrification temperatures,
3. low solubility of noble metals in glass.

Consolidation of the UDS, recovered soluble Tc, and noble metal fission products within a single alloy waste form is expected to approach the

maximum achievable waste loading and minimize the volume of waste, although this remains to be demonstrated. Application of the materials and methodology developed for metallic wastes from Echem treatment of spent sodium-bonded fuel were considered for the Tc-bearing waste streams from both the UREX and Echem processes. Insights drawn from the development of those materials and from binary phase diagrams were used to estimate the capacities of various mixtures to accommodate the GNEP waste streams. Production of separate waste forms for individual streams could increase the individual waste loadings (e.g., the recovered soluble Tc in the UREX+ process), but would increase the total volume of waste.

Cesium/Strontium Waste

There are three broad classes of waste forms that were considered for stabilization of this waste stream: ceramics (including synthetic minerals), glasses, and cements. These three classes were selected because the process used to make the waste forms included in each class is essentially the same and potentially applicable to remote processing of this highly radioactive waste stream. The first process for all three classes is feed evaporation. For ceramics, the processes involve (1) mixing of raw materials: clay or chemicals, (2) firing or heat-treating these materials together with the waste to affect the desired phase assemblage, (3) optional consolidation with a press prior to or after heat treatment, and (4) packaging. For glass the process involves: (1) mixing the feed with additives, (2) feeding the slurry to a melter where it is converted to a molten glass, and (3) casting the molten glass into the disposal package where it solidifies into a glass. For cement, the evaporated feed is mixed with cement forming materials and solidified into a final waste form. Within each class, there is some variation on the overall process for specific materials, but, for the most part, these overall process steps are applicable. For example, in the fluidized bed steam reformer (FBSR) process, Steps 1 and 2 are combined in a steam environment. Further consolidation of the sintered material may be needed.

Metal matrix waste forms are also discussed. However, this option is better considered as a canister and storage strategy than a waste form option, per se, because the waste itself is still fixed first in a glass, a ceramic, or even a simple oxide. These stabilized materials could then be dispersed in a metal matrix to enhance the overall conduction of heat and provide some radiation shielding. This option allows the waste loading in the primary waste form to be much higher than would be possible for the material alone while reducing the centerline temperature of the waste package or allowing a much larger diameter waste form to be stored.

For all of these materials transmutation and decay heat are the major technical materials challenges to resolve. For example, ^{137}Cs decays to ^{137}Ba; ^{90}Sr decays to ^{90}Y, which then decays to ^{90}Zr. With these decays come changes in charge on the cation and ionic radius. The affects of these changes on all the materials considered are largely unknown, although glass is expected to be less sensitive to the change in ionic radius than crystalline materials. It was assumed that the change in charge can be compensated by including in the crystalline or vitreous matrix elements that have one or more than one available oxidation states (e.g., Fe(III) in Fe2O3 can go to Fe(II) and FeO or Fe3O4) with the consumption or release of oxygen. The capacity for charge transfer is found in naturally occurring minerals such as garnet, where Fe(III) and Fe(II) on different crystal sites can exchange charge. It is also assumed that changes in ionic radius can be accommodated without degrading the waste form, but this has not been demonstrated. From a processing point of view, the presence of high concentrations of ^{137}Cs and ^{90}Sr in the waste stream means that careful processing may be needed to handle the high β-γ dose and associated decay heat.

A composite glass-bonded ceramic waste form (CWF) was specifically developed to immobilize radionuclides in chloride salt wastes from the Echem treatment of spent sodium-bonded fuel. During processing, the salt reacts with zeolite 4A to sequester chloride in a sodalite phase, and the sodalite is encapsulated in a borosilicate glass to produce a multiphase glass-bonded sodalite waste form. Most of the radionuclides either dissolve in the glass or form phases that become included in the glass. The principle role of the sodalite phase is to contain the chloride (and iodide). Thus, the waste loading that can be achieved is limited by the amount of zeolite that is needed to sequester the chloride and the amount of binder glass needed to encapsulate the sodalite. Due to the chloride intrinsic to this waste stream, no other options were considered.

Lanthanides and Balance of Fission Products

In both aqueous and electrochemical processing, separation of the lanthanides (Ln) is being developed. In UREX+1a, the non-Ln FP including isotopes of the transition metals Zr, Nb, Mo, Ru, Rh, and Pd are segregated, but could be combined with the lanthanides in the same waste form.

The Echem process flowsheet may leave lanthanide chlorides in waste salt. Use of the CWF for the salt waste stream as described above for Cs/Sr would then be used, and no options were evaluated due to the chloride content intrinsic to this waste. However, if the lanthanides are recovered electrochemically, the salts can be distilled away and the lanthanides could be stabilized using any of the waste forms described in this section. In Echem transition metal FP are retained in the anode basket and then incorporated into a metal waste form as described above, no options were considered here due to the high waste loading and density of this demonstrated waste form.

The candidate waste forms for the combined Ln/FP streams can be grouped into four primary classes: glasses (e.g., borosilicate and phosphate); mineralized forms (e.g., aluminosilicate or phosphate minerals); ceramics (e.g., Synroc-type), and composites (e.g., glass bonded zeolite). Metallic waste forms are also included as potential means to treat fractions of the fission product streams.

Vitrification of the Ln and FP streams would have several advantages, including relatively high loadings, a proven technology, and similarity in form to waste forms currently accepted for repository disposal. Waste loadings of 30-60 wt% for lanthanides have been demonstrated in the laboratory, and 20-30 wt% combined Ln/FP, limited by noble metal solubility, is estimated. Glasses have been developed and tested and fabrication processes have been demonstrated for a wide variety of applications with similar compositions and disposal requirements to the Ln/FP wastes. A second glass option considered was an iron-phosphate glass (IPG). The IPG is an attractive option due to the relatively high solubility of salt components, such as molybdate and sulfate in the phosphate liquid. While most phosphate-based glasses are relatively low in chemical durability and are not suitable for nuclear waste forms, two phosphate-based glass families have shown superior durability: the alkali-alumino-phosphate family and the alkali-iron-phosphate families. In the vitrification process, the feeds will be prepared for vitrification by mixing the feeds with glass frit or appropriate glass forming chemicals, melted in a high temperature melter, cast into a container, and stored.

The primary melter technologies considered include the Joule Heated Melter (JHM) and Cold Crucible Induction Melter (CCIM). Joule-heated ceramic melters are currently used in radioactive operations for treatment of HLW in the U.S. CCIM are currently receiving increased interest due to their ability to process at higher temperatures, minimize melter corrosion by use of a skull layer to contain the melt, and allow processing with significant amounts of crystalline inclusions in the melt.

In a mineralized waste form, various solid mineral phases can be produced depending on the type of co-reactant fed with the waste (e.g., a high sodium waste mixed with an aluminosilicate clay produces a sodium aluminosilicate [NAS] mineral waste form). For the Ln/FP wastes, a number of waste-specific mineralized forms were considered based on the major constituents of the wastes. A FBSR was considered as the primary process to produce mineralized-type waste forms, though this is only one of several reactors that could be used (i.e. a rotary calciner). Waste fed to a steam reformer can be either liquid or slurry. The resulting mineralized product is collected from the reaction vessel. The particulate waste form would probably have to be consolidated in some form prior to disposal.

Ceramic-based (or more appropriately, crystalline) waste forms retain the radionuclides in the waste as part of the matrix. The primary ceramic-based materials evaluated were those from the Synroc and monazite families of compositions. In general, Synroc (i.e., synthetic rock) is an advanced synthetic crystalline ceramic comprised of geochemically stable titanate-based minerals, which have immobilized uranium, thorium, and other naturally-occurring radioactive isotopes in the environment for millions of years. These minerals and their man-made analogs are capable of incorporating into their crystal structures nearly all of the transition metal FP and the

Ln/FP. Monazite is also attractive as a form based the fact that significant amounts of the naturally occurring actinides thorium and uranium are contained in natural monazites. Ceramic materials can be formed using a wide range of processes including cold pressing and sintering, hot uniaxial or isostatic presses, and CCIM. Of these various processing routes, HIP is the most developed for production of Synroc-type materials. Throughput limitations have hampered the utility of ceramic processes for large-scale waste treatment operations. The use of the CCIM technology to produce ceramic forms may provide a solution to this limitation.

Volatile Radionuclides

For the purposes of this study it was assumed that the primary volatile radionuclides from a large reprocessing facility could not be vented into the atmosphere or discharged to water bodies. Once captured, ^{129}I and ^{14}C must be sequestered essentially indefinitely, but ^{3}H, ^{85}Kr can be effectively managed in decay storage due to their relatively short half-lives (12.28 years, 10.73 years, respectively). Proposed capture methods were evaluated using parameters such as selectivity, efficiency, regeneration of sorbent, and conversion to final waste forms. Silver-zeolite (AgZ) for iodine, molecular sieve for tritium, caustic scrub for ^{14}C, and zeolite (mordenite, faujasite) for Xe/Kr are the baseline technologies selected in the conceptual design of an offgas control system.

When loaded with tritium-bearing water, the molecular sieves may be added to grout for final disposal or regenerated by desorbing the water. A reasonable disposal path includes adding either the loaded molecular sieves or the recovered water to grout, placing the grout in a stainless steel drum, sealing the drum, and disposing of the drum by burial. The relatively short half-life of tritium (12.26 years) ensures that it will decay to safe levels before the packaging will deteriorate. Grouting is a well-developed technology for stabilizing a variety of waste forms. No problems are anticipated for this method of disposing of a purified tritium bearing water stream. However, because the molecular sieves may also co-sequester a small amount of ^{129}I and carbon dioxide (^{14}C) the method should be evaluated for the effect on a grout waste form and options should be identified and/or developed to cleanly separate the water from these other species.

A number of possible immobilization forms have been suggested for iodine. The dissolution of NaI or KI in standard fluoride glasses is limited to 1 mol%. Quarternary glasses permit compositions of 4 and 8% iodide respectively, but the durability of these glasses is quite low. One possible stabilization package is based on the incorporation of the iodine-loaded AgZ into a grout matrix. Assuming that the iodine "filter" is designed as the storage package, then this would have end caps welded in place and the sealed filter package would be placed into a secondary overpack. Grout could be added to the annular space, but this would add little to the overall containment of the ^{129}I. A second approach is to remove the loaded AgZ pellets from the filter housing and mix the loaded pellets with grout for stabilization and containment during transport. A disposition pathway for this waste form considering the 1.6×10^{7} year half-life of ^{129}I in something other than a vitrified HLW form is yet to be resolved. A number of studies have also shown that iodine-loaded silver zeolites can be converted to the aluminosilicate mineral sodalite in which the iodine is more strongly bound than in the unprocessed zeolite sorbent. Iodosodalite that does not contain silver has also been successfully synthesized and tests indicate that it may be suitable as a long-term waste form.

Options evaluated for Kr include storage in pressure containers and encapsulation in solid matrices. Pressurize gas containers must remain intact for ~100 years; resist corrosion due to the in-growth of elemental Rb that is chemically aggressive; and dissipate the decay heat. This method provides for easy recovery of the krypton for subsequent industrial use, but it also increases the hazard of a release. Encapsulation as a sputtered metal matrix will contain 5–6% Kr on an atomic level. The product is an amorphous glassy deposit. Depending on the process used, loading of 16–25 liters at standard temperature and pressure (STP) per kg metal matrix could be achieved. An alternative is encapsulation in a zeolite matrix. The krypton is encapsulated in the zeolite structure by a sintering process where the pores of the zeolite are sealed. The relatively low thermal conductivity of the zeolite should be considered and may limit the maximum loading of the zeolite.

Most of the carbon immobilization studies conducted to date have considered calcium or

barium carbonate that has been mixed with cement and packaged in steel drums.

COMPARISON OF WASTE FORMS AND TECHNOLOGIES

Comparison of waste forms and technologies to form a baseline was complicated by the wide variety of radionuclides considered and the differences between aqueous and electrochemical waste streams. For example, both reprocessing concepts release tritium, but iodine and carbon are released during aqueous dissolution, but retained in the molten chloride salt used by Echem. Also, some judgments are very subjective; powder processing is used commercially every day to fabricate fuel, but it may be unacceptably problematic to consider for remote production of intensely radioactive Cs/Sr waste forms. With those limitations in mind, the processing options were evaluated based on the parameters in Table 3. Waste treatment experts from across the U.S. were enlisted to evaluate the options using these criteria with only slight modifications to fit the wastes in consideration.

TABLE 3. Waste Form Evaluation Criteria.	
Commercial Practicability (weighted 40%)	
	Technical Practicality
	Flexibility/Robustness
	Complexity of Process
	Scalability
	Waste Loading
	Processing Cost
	Transportability
	Secondary Waste Generation
Technical Maturity (weighted 10%)	
	Development Cost
	Schedule to Implement
	Process/Product Maturity
Waste Form Performance (weighted 40%)	
	Thermodynamic stability
	Rad/Mech/Thermal Stability
	Chemical Durability (e.g., TCLP, PCT)
	Predictable Performance
Waste Stream Specific Criteria:	
	Heat transfer and degradation
	RedOx Sensitivity
	Resistance to degradation by decay, valence change, atomic size, chemistry
	Resistance to radiolysis effects gas generation from water, degradation of the waste form
	Ability to treat more than one waste
	Experience with disposal of similar materials
Stakeholder Acceptance (weighted 10%)	
	All processes designed to meet standards

RESULTS

The recommended baseline for the GNEP waste streams are summarized in Table 4. Note that options are retained in several instances pending process development. Waste forms recommended are based on current assumptions, including the general UREX+1a and Echem process flowsheets, current U.S. regulations, and the overriding assumption that the text in the Nuclear Waste Policy Act will be reconsidered such that the “highly radioactive material resulting from the reprocessing of spent nuclear fuel” will not all have to be disposed as HLW in a geologic repository by law, but disposal requirements for the separate waste forms can be evaluated on their own merits.

CONCLUSIONS

This summarizes a much more detailed study for selecting the most technically affective waste forms for stabilizing the primary waste streams from GNEP for their planned disposition as summarized in Tables 1 and 4. In most cases, recommendations were made based on insights from existing data available for similar materials, usually by researchers having extensive experience with those materials. However, little if any data exists for several of the envisioned waste forms, including if they can be made using the waste stream, if the expected waste loading can be achieved, the impact of contaminants, and how practically the manufacturing processes can be engineered for application in a hot-cell. Results from the recommended R&D may well result in other waste forms being recommended. Additional regulatory analyses are needed and process engineering analysis will almost certainly affect these recommendations. Some processes, particularly the conveying, manipulating, and containing of highly radioactive powders and self heating solutions, are not readily designed for implementation in a remote environment. Although those factors were taken into account in selecting the baseline waste forms, more detailed facility engineering studies could lead to different assessments. Waste acceptance criteria for disposal facilities, many of which do not yet exist, will also affect these recommendations.

TABLE 4. GNEP Waste Form Baseline Recommendations.		
Waste Stream	Waste Form(s)	
	UREX+1a	Echem
Tc	Metal alloy, possibly containing UDS and transition metal FP. Alloy may require added Zr or Fe, which could be provided by cladding and hardware.	Metal alloy containing UDS and transition metal FP. Alloy may contain cladding, and may require supplemental Zr or Fe, which could come from additional cladding and hardware.
Cs/Sr	Glass or Ceramic, process design should consider ramifications of high heat, high radioactivity, powder handling should be avoided.	Glass-bonded sodalite.
Ln	Lanthanide – high loading Ln-glass if segregated as separate Ln stream. Ln/FP borosilicate glass if Ln and FP streams are combined.	Lanthanide - high loading Ln-glass if segregated as separate Ln stream. Glass-bonded sodalite if Ln are in salt.
FP	Metal alloy potentially combined with Tc and UDS. Borosilicate glass if combined with lanthanides	Metal alloy containing Tc and transition metal FP. Matrix may contain cladding, and supplemental Zr/Fe could come from additional cladding and hardware.
UDS	Metal alloy potentially combined with Tc and FP.	Metal alloy containing Tc and transition metal FP. Matrix may contain cladding, and supplemental Zr/Fe could come from additional cladding and hardware.
Metals—Cladding/Hardware	Compacted metal. Metal ingot if cost effective.	Compacted metal. Metal ingot if cost effective.
Tritium	Grouted tritiated water (HTO).	Grouted tritiated water (HTO).
Iodine	Heat treated or grouted silver zeolite.	Glass-bonded sodalite w/Cs/Sr and possibly Ln.
Carbon-14	Grouted Na/CaCO3.	Glass-bonded sodalite w/Cs/Sr and possibly Ln
Krypton	Pressurized gas cylinder w/wo Xe.	Pressurized gas cylinder w/wo Xe.

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